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STRUCTURE AND DYNAMICS OF EXCITED ATOMS

Final Report Grant No. F49620-96-0014

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by

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I. Introduction

With the support of AFOSR grant F49620-96-1-0014 we have examined two topics, resonant energy transfer between Rydberg atoms and microwave multiphoton processes in Rydberg atoms. In both of these lines of research we take advantage of the exaggerated properties of Rydberg atoms to explore physical regimes which would be hard to study quantitatively in other systems. For example, in the experiments with cold Rydberg atoms we have seen the evolution from two body collisions to many body interactions as we increase the strength of the interaction between atoms.

II. Resonant Energy Transfer

Using an atomic beam we have examined in detail the collisional energy transfer

$$K 26s + K 24d \rightarrow K 26p + K 25p,$$
 (1)

which is resonant at several fields from 20 to 26 V/cm. Specifically, we have used different distributions of the velocities of the colliding atoms. When the velocity distribution is as wide as the average velocity, the resulting lineshape is cusp shaped. Thus the velocity distributions for thermal atoms in a cell or an atomic beam give cusp shaped lines. Similarly, collisions between atoms in a single velocity group give a cusp shaped line. In contrast, collisions between atoms in two distinct velocity groups lead to Lorentzian lineshapes.

Much of our effort during this period was focused on the development of a magneto optical trap for the study of the interactions of cold Rb Rydberg atoms.¹ Using the trap we have been able to produce Rb densities of 10¹⁰ cm⁻³ in a volume of 10⁻³ cm³, and we have studied several collision processes. The one we have examined in most detail is

$$Rb 33s + Rb 25s \rightarrow Rb 34p + Rb 24p \tag{2}$$

which is resonant at 3 V/cm, and we have observed resonances as narrow as 3 MHz. In the cold Rydberg gas the atoms move $\sim 10\%$ of their typical spacing in 3 μ s, the time of the experiment, so the energy transfer is not due to a collision but to the static interaction of the atoms. In other words, we have created an artificial solid.

III. Microwave Multiphoton Processes

During the grant period we examined microwave multiphoton processes involving both ionization and transitions between bound states.

A subject of great theoretical interest is ionization by circularly polarized fields,^{2,3} and we have reported the ionization of both Na and Li by 8 and 18 GHz circularly polarized fields. In the former experiments we have demonstrated that even at a frequency of 18 GHz the ionization field is well described by

$$E = 1/16n^4$$
, (3)

in marked contrast to a theoretical estimate which is far lower.² We have measured the sensitivity to ellipticity of the polarization and developed a simple, quantitative model to account for the observations. Finally, we have shown that small static fields in the plane of the rotating microwave field produce striking enhancements in the ionization. For both elliptical polarization and an applied static field the enhancement originates from resonant transitions between the rotating frame eigenstates driven by the static field or the counter rotating component (for elliptical polarization).

In the Li experiment we had hoped to see a striking difference between the $\ell+m$ even and $\ell+m$ odd states, because the latter are much more hydrogen like. Ionization of Na m = 2 states by linearly polarized fields is very different from that of the m = 0 and 1 states for this reason.⁴ However the ionization of both sets of states occurs at essentially the same field, a somewhat disappointing, but nonetheless enlightening, result. An additional surprising result of the experiment was our ability to see clear structure in the excitation spectra of the $\ell+m$ odd states, excited via the np m = 0 levels, but none in the $\ell+m$ even states excited via the np m = ±1 levels. The difference is due to the energy shift of +m ω in transforming between the laboratory and rotating frames.⁵ For m=0 there is no shift, but the m = ±1 states, which are degenerate in the laboratory frame, are split by 2ω in the rotating frame of the field, doubling the number of observed lines and blurring the resulting spectrum.

One of the more interesting questions about microwave ionization is whether or not all atoms look like H if the time is short enough, and we have examined the ionization of Na by 600ps 8 GHz pulses. The Na atoms clearly behave like H, and for the first time we have seen population transfer to other bound states.

Using rf pulses of frequencies between 400 and 600 MHz we have examined how a two level system, composed of K Rydberg states, responds as the rf pulse is changed from one to many cycles. The objective is to connect recent half cycle pulse results⁶ to conventional Floquet descriptions of resonance phenomena.⁷ With one cycle we observe a fairly structureless response. With two cycles what was an unstructured response with one cycle exhibits interference fringes much like a Young's two slit interference With progressively more cycles the interference maxima develop into sharp resonances. Furthermore, we have shown that in the perturbation theory limit the observed resonance pattern is identical to a diffraction pattern. In the course of working out the theory connecting the one cycle response to the many cycle response we worked out a form of Floquet theory which was first developed by Moloney and Meath⁸ twenty years ago and then largely ignored. This form of Floquet theory requires integration over a complete field cycle, but allows any shape of cycle, not only sinusoidal variation. It is a very powerful approach and leads to some remarkable insights. For example, it predicts that for multiphoton transitions even if the oscillating field is present for thousands of cycles, how the first cycle turns on is absolutely crucial. We have done experiments to verify this prediction, and we have shown that there is a clear difference between a pulse which is an integral number of cycles of sine and cosine. The commonly used form of Floquet theory is an average of the sine and cosine pulses, which gives a flawed description of pulses which turn on in more than one cycle. Such pulses are almost identical to sine pulses.

We demonstrated how to produce square wave Rabi oscillations by frequency modulating the resonant coupling field, and showed how it could be easily understood as adiabatic rapid passage between dressed states.

IV. Stark Avoided Crossings

Since we are using Rb for the trap experiments, we need to know the spectroscopy in E fields and understand field ionization in Rb. For both of these reasons we have measured the avoided level crossings of the Rb (n+3)s states with lowest levels of the adjacent n Stark manifolds. Our measurements agree with values obtained from numerical calculations using the coulomb approximation, and we were able to develop a perturbation theory expression showing the dependence on the quantum defects of the s states as well as well the n dependence.

V. Publications

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VI. Conclusion

During the grant period we have started experiments in two areas; cold trapped Rydberg atoms, and short microwave pulses, which offer the promise of many new insights. We expect to realize these over the next few years.

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